Anomalous temperature dependence of magnetic quantum oscillations in CeBiPt

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Shubnikov–de Haas (SdH) and Hall-effect measurements of CeBiPt and LaBiPt reveal the presence of simple and very small Fermi surfaces with hole-like charge carriers for both semimetals. In the magnetic material, CeBiPt, we observe a strong temperature dependence of the SdH frequency. This highly unusual effect might be connected with an internal exchange field within the material and a strongly spin-dependent scattering of the charge carriers.

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The Shubnikov–de Haas (SdH) effect, i.e., quantum oscillations in the magnetoresistivity $\rho(B)$, was discovered about 70 years ago in the semimetal Bi [1]. Since then, innumerable studies on the SdH effect and the related de Haas–van Alphen (dHvA) effect in the magnetization were reported for many metals, metallic compounds, and semimetals. Both effects arise from the oscillations of the free energy of the electrons as with increasing field B a sudden change of population of Landau cylinders occurs whenever a Landau cylinder is pushed through an extremal cross-section A of the Fermi surface perpendicular to the magnetic field. The oscillations are periodic in $1/B$ and the frequency F is directly proportional to A, i.e., $F = (\hbar/2\pi e)A$. The theory of the dHvA effect is well established [2], and also the SdH effect, reflecting the more involved transport properties, is principally understood [3]. In recent years, SdH and dHvA effects have been exploited to establish the Fermi surface of complex materials with strong electron correlations such as heavy-fermion systems [4]. In many cases, the effective mass of the carriers in a given band of the Fermi surface can be determined from the temperature dependence of the amplitude of the oscillations. Here we report on a new observation, namely a strongly temperature-dependent *frequency* of the SdH oscillations. This anomalous T dependence of F was found for a certain field orientation in semimetallic CeBiPt, while it is absent in the homologous - except for the Ce $4f$ electron - semimetal LaBiPt.

The single crystals were grown at Hiroshima University with the Bridgman technique in a hermetically sealed Mo crucible from the starting materials Ce (m5N, Ames Laboratory), Bi (m5N), Pt (m3N), and La (m4N). To avoid oxidation of elemental Ce or La during handling, CePt or LaPt were first prepared by argon-arc melting and the appropriate amount of Bi then was added for single-crystal growth. The crucible, sealed under Ar atmosphere, was heated to 1350°C in a furnace (with an intermediate halt at 500°C for 2 h) and after 12 hours was slowly cooled by moving it out of the central zone of the furnace with 1 mm/h. This cooling process from 1350°C to 20°C took 6 days, then the furnace was switched off. Whether the 1-1-1 compounds or 3-4-3 compounds like $Ce_3Bi_4Pt_3$ are formed, depends very sensitively on the excess amount of Bi. The 1-1-1 compounds show the $F\bar{4}3m$ cubic structure previously determined for polycrystalline samples [5] with no indications of second phases. The lattice constant $6.8338\,(25)$ Å found for CeBiPt is in good agreement with the previously reported value [5]. For the LaBiPt crystal, Laue-diffraction pictures showed some mosaicity of the sample.

The longitudinal and transverse magnetoresistance (Hall effect) were measured with a standard ^{3}He cryostat setup up to 15 T in Karlsruhe, and up to 28 T at the High Magnetic Field Laboratory in Grenoble. Both sets of data agree with each other in region of overlap. Six gold wires were glued with graphite paste to the samples, thereby enabling to measure simultaneously the longitudinal (ρ_{xx}) and transverse (ρ_{xy}) magnetoresistance. These resistances were measured by use of a low-frequency ($\sim 16\,$Hz) lock-in technique for normal and reversed field orientations which allowed a well-defined separation of ρ_{xx} and ρ_{xy}. The dHvA signal of the LaBiPt sample was measured by means of a capacitance cantilever torque magnetometer. All sample holders could be rotated *in situ* around one axis.

Fig. 1(a) and (b) show the magnetoresistance $\rho(B)$ for CeBiPt and for LaBiPt with field along [1 0 0] for several temperatures. The current direction was perpendicular to B. In both cases, clear oscillations are seen. For LaBiPt at $T = 0.4\,$K, the strong decrease of ρ below about 1 T (with a 50% value at 0.45 T) indicates the onset of superconductivity with $T_c = 0.88\,$K [see inset of Fig. 1(b) for $\rho(T)$]. The rather large critical-field slope $(\mathrm{d}B_{c2}/\mathrm{d}T)_{T_c} \approx$ -1 T/K hints at a small coherence length. For some field directions we observed a beat pattern for LaBiPt which might be attributed to the mosaicity mentioned above. We can, however, not exclude the existence of a second extremal Fermi surface of small area in

LaBiPt.

For CeBiPt, a first unusual observation is the sharp drop of the magnetoresistance at low fields and low temperatures [Fig. 1(a)]. Below about $T_N = 1\,$K, CeBiPt is in an antiferromagnetically ordered state as was evidenced by specific-heat and magnetization measurements for our samples [6,7]. Below T_N, the field-dependent magnetization, M, has a maximum in $\mathrm{d}M/\mathrm{d}B$ at about $0.3\,$T [6]. Therefore, the negative magnetoresistance at low fields presumably is caused by antiferromagnetic fluctuations which become reduced in an applied magnetic field.

The most striking observation for CeBiPt is the shift of the oscillating SdH signal with temperature. This can be made more apparent when the resistivity is converted to conductivity via $\sigma_{xx} = \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2)$ making use of the simultaneously measured transverse magnetoresistance ρ_{xy} [8]. A smooth background longitudinal conductivity σ_0 can be fitted to the σ_{xx} data to obtain the SdH signal $\Delta\sigma/\sigma_0 = (\sigma_{xx} - \sigma_0)/\sigma_0$ as shown in Fig. 2. The inset of Fig. 2 displays the sequence of the position of $1/B$ of the oscillation maxima and minima.

The oscillations are indeed periodic in $1/B$ within our resolution and for the limited field range. The oscillation frequency F is directly obtained from the slope of $1/B$ vs. oscillation index n. F decreases from $58.2\,$T at $0.4\,$K to about $35\,$T at $10.3\,$K. Additional data taken in Karlsruhe in fields up to $15\,$T are fully in line with this decrease (see Fig. 3). The total change of F between 0.4 and $10.3\,$K corresponds to an apparent reduction of the Fermi-surface cross-section by more than 50%! The inset of Fig. 3 shows the decrease of the amplitude A of the SdH oscillations with increasing T as measured at $10.5\,$T. A can be described quite well with the standard expression $A \sim T/\sinh[(14.69(T/B)(m^*/m_0))]$, where T and B are expressed in K and T, respectively [9]. We obtain the effective mass $m^* = 0.24\,m_0$, where m_0 is the free-electron mass. A simple consistency check serves to test whether we are indeed dealing with a quantum-oscillation phenomenon. From the field dependence of $\Delta\sigma/\sigma_0$ at fixed T one can estimate the charge-carrier scattering rate τ^{-1}. The Dingle temperature $T_D \approx 4\,$K obtained from the field dependence of the SdH amplitude (at lower fields) corresponds to $\tau^{-1} = 3.3 \times 10^{12}\,\mathrm{s}^{-1}$. Together with the carrier density $n_h = 7.7 \times 10^{17}\,\mathrm{cm}^{-3}$ obtained from the Hall effect at liquid-He temperature we obtain in the simple Drude model $\rho \approx 3.7\,\mathrm{m\Omega cm}$ which is perfectly in line with the measured resistivity.

The angular dependence of F for CeBiPt is depicted in Fig. 4(a) for $T = 0.43\,$K and $4.21\,$K. The anomalous T dependence is found only for field along the $[1\,0\,0]$ and $[0\,1\,0]$ directions where F is found to be somewhat larger, i.e., about $58\,$T. Between approximately $\theta = 20°$ and $70°$, where θ is the angle from the $[1\,0\,0]$ direction, F is constant at $30\,$T and furthermore independent of T. For B along the $[1\,1\,1]$ direction a very low SdH frequency of about $20\,$T - but again independent of T - was found. LaBiPt shows a temperature-independent angular dependence of F [Fig. 4(b)]. The SdH and the dHvA frequency are within error bars identical and somewhat larger (F increases from $65\,$T for B along $[1\,0\,0]$ to $95\,$T for B along $[1\,1\,0]$) than F for CeBiPt. The large magnetization of CeBiPt due to the Ce $4f$ electrons prevented us from taking dHvA data of the torque magnetization for this compound. Nevertheless, the very good agreement between SdH and dHvA data for LaBiPt lends support to the SdH data for CeBiPt. For both materials we were able to observe a SdH signal over the whole angular range. This suggests that the Fermi surfaces are simple single-connected hole pockets. The volume enclosed by the Fermi surface is estimated to comprise only about 1.5×10^{-4} of the volume of the first Brillouin zone for CeBiPt. This is consistent with the low (hole-like) charge-carrier concentration of $n_h = 7.7 \times 10^{17}\,\mathrm{cm}^{-3}$. Within a free-electron picture, n_h corresponds to a Fermi energy of $E_F = \hbar^2(3\pi^2 n_h)^{2/3}/2m^* = 12.8\,$meV. Assuming a circular Fermi-surface cross section, i.e., $A = \pi k_F^2$ with the Fermi wavevector k_F, this results in a SdH frequency of $F = m^* F/\hbar e \approx 27\,$T in nice agreement with the experimental F between $20°$ and $70°$.

The main point of the present investigation is the observation of a temperature dependence of the quantum-oscillation *frequency* which is found only for the Ce-based metal. To our knowledge such an effect has never been observed before. As a possible origin for this behavior one could assume a temperature-dependent charge-carrier density which would lead to a change in the Fermi-surface topology. However, the simultaneously measured Hall constant $R_H = 1/n_h e$, is independent of temperature.

Another possibility would be a reconstruction of the Brillouin zone. This might be caused, e.g., by an antiferromagnetic ordering, which introduces an extra periodicity into the lattice. The effect of magnetic ordering has previously been observed in a *field-dependent* change of the dHvA frequency in NdIn$_3$ [10] or in an unusual temperature dependence of the dHvA *amplitude* in YbAs [11], SmSb [12], and SmAgSb$_2$ [13]. As mentioned, CeBiPt undergoes an antiferromagnetic transition at about $1\,$K. However, no abrupt change of F with T around T_N or B but a rather smooth variation over a large T range is observed. This indicates that a Brillouin-zone reconstruction must be ruled out as a cause for the observed frequency change.

One important fact which is evident from our investigation is the absence of any unusual effect for the non-magnetic sister compound LaBiPt. Therefore, it is clear that the magnetism of the Ce atoms affects the magnetic quantum oscillations. For a sample possessing an internal magnetization, the magnetic induction $B_i = \mu_0(H + M)$ is different from the externally applied field $B = \mu_0 H$. The SdH signal is proportional to

$\sin(2\pi F/B_i)$. Therefore, a temperature-dependent magnetization would cause a change in the SdH frequency. However, in order to explain the experimentally observed increase of the SdH frequency with decreasing temperature, M would have to become smaller at lower temperatures. This is contrary to the usual behavior of M and not in line with the measured magnetization which is about $\mu_0 M = 0.2\,\mathrm{T}$ at $\mu_0 H = 12\,\mathrm{T}$ and $T = 1.7\,\mathrm{K}$ and decreases with increasing temperature. Moreover, the magnitude of $\mu_0 M$ compared to B is much too small to account for the observed change of F.

An anomalous situation may occur when we suppose that a temperature-dependent (and possibly also a field-dependent) phase difference Φ exists between the spin-up and spin-down oscillations and that, in addition, their amplitudes may be different [9]. The latter can easily occur because the scattering by magnetic impurities (or magnetic sublattices) is in general appreciably spin dependent. The usual splitting of the spin-up and spin-down Landau levels gives rise to a phase shift of the oscillations by $\Phi = \pm\pi g m^*/m_0$, where g is the g factor of the conduction electrons. If an antiferromagnetic exchange interaction B_{ex} is present, the phase difference can be expressed as $\Phi = \pm\pi(g - B_{ex}/B)m^*/m_0$ [9]. The superposition of the spin-up and spin-down oscillations gives a signal which is $M = a_\uparrow \sin(\psi + \Phi/2) + a_\downarrow \sin(\psi - \Phi/2)$, with $\psi = (2\pi F/B) \pm \pi/4$ and the spin-dependent amplitudes $a_\uparrow$ and $a_\downarrow$. In case of largely different amplitudes, the frequency of the oscillating signal becomes $F' = F \pm B_{ex}/4$. Adopting this scenario would, however, imply a change of B_{ex} with T by several $10\,\mathrm{T}$ which appear unlikely. On the other hand, the apparent decrease of the oscillation frequency in $1/B$ above about $0.12\,\mathrm{T}^{-1}$ at low T does suggest an influence of internal and/or exchange fields. Calculations of spin splitting in low-carrier-density systems are necessary to check the viability of this scenario.

In summary, we have presented an unusual T dependence of SdH oscillations in CeBiPt which are due to the magnetic Ce $4f$ electrons. Simple estimates of the scattering rate derived from the Dingle temperature and of the volume enclosed by the Fermi surface yield good agreement with the corresponding quantities obtained from other measurements. This lends confidence to the assignment of the observed apparent T dependence being a feature associated with the magnetic quantum oscillations in CeBiPt. However, theoretical work is needed to unravel the origin of this new phenomenon.

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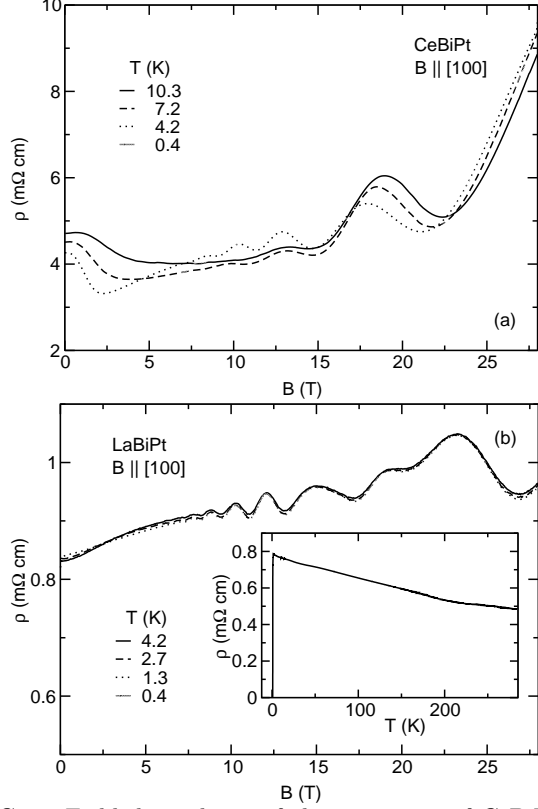
FIG. 1. Field dependence of the resistivity of CeBiPt (a) and LaBiPt (b) for selected temperatures. The inset shows the temperature dependence of the resistance of LaBiPt with the superconducting transition at about 0.88 K.
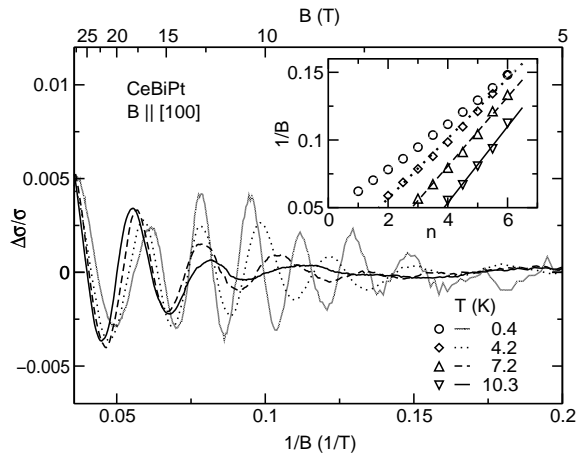

FIG. 2. Field dependence of the SdH signal, i.e., the relative conductance oscillations, of CeBiPt for different temperatures. The inset shows the peak and valley positions vs an arbitrary index n.
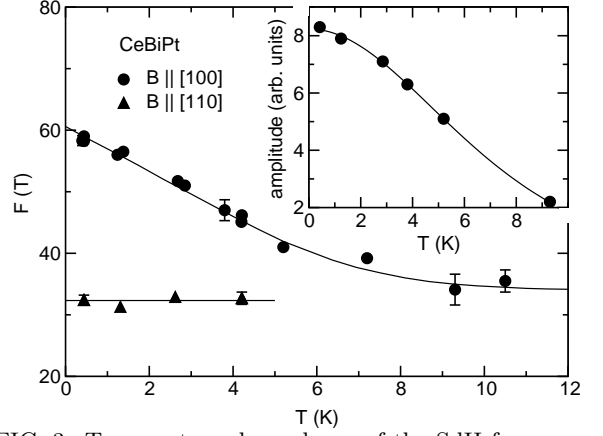

FIG. 3. Temperature dependence of the SdH frequency of CeBiPt for two different field orientations. The lines are guides to the eye. The inset shows the temperature dependence of the SdH oscillation amplitude with the theoretical fit for an effective cyclotron mass of $m^* = 0.24\, m_e$.
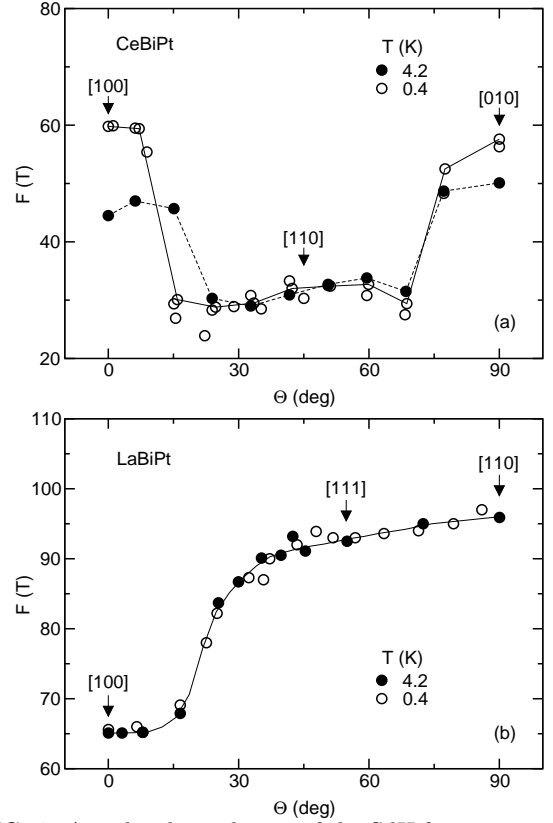

FIG. 4. Angular dependences of the SdH frequencies of (a) CeBiPt and (b) LaBiPt for two different temperatures.